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SUPERCONDUCTING ELECTRONIC FILM STRUCTURES

A. I. Braginski, J. R. Gavaler, and J. Talvacchio
Superconductor Materials and Electronics

Semiannual Report for the Period
January 1 to June 30, 1988

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application. Y-Ba-Cu-O/MgO/Nb tunnel junctions were fabricated. Non-continuous barrier layer produced superconducting shorts in the junctions. This result, however, provided the non-ambiguous evidence that the new growth procedures had produced films which were superconducting up to the surface.

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1. SEMIANNUAL REPORT, SUPERCONDUCTING ELECTRONIC FILM STRUCTURES

January 1, 1988 to June 30, 1988

AFOSR Contract No. F49620-88-C-0039

A. I. Braginski, J. R. Gavaler, and J. Talvacchio

2. ABSTRACT

Modifications in Y-Ba-Cu-O growth procedures were instituted to eliminate non-superconducting near-surface layers observed in films. These include much higher ramp rates to the 850°C crystallization temperature of the as-deposited amorphous films and the use of an entirely *in-situ* processing. Films have now been grown which are superconducting up to the surface. Other Y-Ba-Cu-O films have been sputtered which are crystalline (tetragonal) as-deposited. These films were shown to be more stable at high temperatures than amorphous films. Gold layers deposited at room temperature on recent films were found to have contact resistance less than the limit of the apparatus sensitivity. Evidence for crystallinity and epitaxy was found in 10 nm YBCO films deposited at 650°C from the vapor phase. Measurements on superconducting resonator cavities have begun using epitaxial films on SrTiO₃ substrates. Thin epitaxial MgO layers were grown on sapphire to eventually provide a better substrate for this application. Y-Ba-Cu-O/MgO/Nb tunnel junctions were fabricated. Non-continuous barrier layer produced superconducting shorts in the junctions. This result, however, provided the non-ambiguous evidence that the new growth procedures had produced films which were superconducting up to the surface.

3. OBJECTIVES

The objectives of the Westinghouse-AFOSR program are:

1. Investigate methods of high- T_c oxide film growth.
2. Search for optimal tunneling barriers, encapsulation, and metallic contact materials compatible with high- T_c oxide superconductors.
3. Grow epitaxial high- T_c oxide films and coherent, layered film structures incorporating tunneling barriers.
4. Study tunneling into high- T_c oxide films and other pertinent electronic properties of films.
5. Develop and evaluate new fabrication and device concepts exploiting the unique properties of high- T_c oxide superconductors.

4. ACCOMPLISHMENTS

4.1 PREAMBLE

The research reported here was done under a Westinghouse-AFOSR Program which began in January 1988. The overall objective of this program is to fabricate and characterize layered film structures incorporating high-critical-temperature oxide superconductors in a manner permitting electronic device applications. Components fabricated from these film structures - especially, tunnel junctions (if feasible) and transmission lines - will form a base for the technology of superconducting electronics capable of operating at or above 50K. Studies performed during the period of January 1, 1988 to June 30, 1988 are described in this report.

4.2 HIGH- T_c OXIDE FILM GROWTH

Work to date has concentrated on thin films of the high-transition temperature (T_c) oxide superconductor, $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO). This compound was chosen as the best representative of the rare-earth barium cuprates. Its properties, both in bulk and in thin film form, have been better characterized than those of any of the other high- T_c superconductors. Films of YBCO are routinely deposited for this program by co-sputtering from three elemental metal sputtering targets. The films are deposited at $<400^\circ\text{C}$ as amorphous oxides. They are then annealed at 850°C in an atmosphere of oxygen to form the superconductor. The apparatus and general procedure for fabricating YBCO films were described in the December, 1987 issue of *Applied Physics Letters*.

When discoveries of the new superconductors, Bi-Sr-Ca-Cu-O and Tl-Ba-Ca-Cu-O, were announced in January and March of this year, some preliminary investigations were conducted with bulk samples of these compounds. From this work, it was concluded that the new materials would not have a fundamentally different impact than YBCO on

applications of high- T_c superconductivity. Therefore, the decision was made to continue to focus thin film work on the better-understood YBCO system. From the work that was performed on bulk specimens, a comparison of transport and magnetization critical currents in Bi-Sr-Ca-Cu-O was made. This comparison showed weak coupling between grains similar to that found in YBCO. The measurements were the first of this type made on this superconductor. They were presented at the March APS Meeting and subsequently published in *Materials Letters*.

As described in the 1987 AFOSR Annual Report, the procedures initially used to prepare YBCO films resulted in films which had a thick non-superconducting surface layer. Two major changes in experimental procedure have since been instituted to improve the surface quality of the films. Through Auger depth profiling and XPS analyses of film surfaces, it was determined that Ba segregated to the surface during the annealing of the amorphous as-deposited films. This segregation occurred at temperatures below the crystallization temperature and resulted in the formation of a Ba-rich non-superconducting layer with a thickness much greater than the YBCO coherence length. It was found that a high ramp rate up to the annealing temperature, approximately $10^\circ\text{C}/\text{sec.}$, minimized this Ba segregation. This work was presented at the Spring Meeting of the Materials Research Society and published in the proceedings.

The second major change required modifications in the deposition system which would permit in-situ annealing in one atmosphere of pure oxygen at temperatures up to 1000°C . Details of the necessary modifications to heaters and the selection of oxygen-resistant materials are included in an invited paper presented at the 1988 Society of Photo-Optical Instrumentation Engineers (SPIE) Meeting on "High T_c Superconductivity: Thin Films and Devices" and will be published in the proceedings. The resulting capability for completely in-situ film processing eliminated degradation of film surfaces due to exposure to air. This capability together with minimization of Ba segregation through the use of high ramp rates now make possible the growth of

multilayers with clean interfaces. The investigation of film surface properties needed for tunneling and device fabrication can now proceed.

One inherent disadvantage still remains when using this particular growth process. That is the relatively high (850°C) temperature that is required to crystallize the amorphous as-deposited films. At this temperature, YBCO reacts with the substrate material to form a degraded interface layer. For SrTiO_3 , the thickness of the degraded layer is approximately 100 nm, as measured by Auger spectroscopy depth profiles and transmission microscopy. Efforts to reduce the overall fabrication temperature by depositing crystalline YBCO films at 600°C have been made. These results were presented at the March APS Meeting. Films were sputtered at 600°C which were crystalline (tetragonal) as-deposited. When annealed at 850°C in oxygen, it was found that these as-deposited crystalline films were more stable at high temperatures than the amorphous films. Auger depth profiles indicated that the thickness of the degraded interface layer had been reduced to less than 10 nm. For the case of epitaxial YBCO films, a comparison between the properties of films crystallized from the amorphous solid state and films crystallized from the vapor phase will be presented at the Applied Superconductivity Conference.

4.3 MATERIALS COMPATIBLE WITH HIGH- T_c OXIDE SUPERCONDUCTORS

During the course of this and also the preceding Westinghouse-AFOSR programs, a large number of materials have been studied with respect to their compatibility with YBCO. At or near room temperature, results have been obtained which indicate that the reactivity of YBCO should present no major problems. For example, gold contact layers were deposited at room temperature on YBCO which showed a contact resistance of less than the limit of the apparatus sensitivity i.e., $<4 \times 10^{-10}$ ohm-cm². These contacts were made on YBCO films with good surface quality which were processed using the in-situ methods described in Section 4.2. This result thus demonstrates that the high contact resistances previously obtained with gold contacts on YBCO at low temperatures were due to the poor surface quality of the YBCO rather

than to any interaction between the metal and the film. A review of the physics and materials science associated with contacts to superconductors will be published in *IEEE Transactions on Components, Hybrids, and Manufacturing Technology*.

Unlike the materials used for contacts and for encapsulation, tunnel barriers may require processing temperatures very much greater than room temperature. It has been already established that typical barriers used in tunnel junctions, such as Al_2O_3 , interdiffuse strongly with YBCO at 850°C . To determine the maximum processing temperature that could be used with an Al_2O_3 barrier, a series of $0.3\text{ }\mu\text{m}$ thick YBCO films were deposited on sapphire and then annealed for various lengths of time and at various temperatures less than 850°C . For films heated slowly and annealed for one hour, a sharp maximum in T_c versus annealing temperature was observed at 820°C . Lower annealing temperatures produced films which were not superconducting. However, films heated rapidly and annealed for just three minutes had T_c 's that were independent of annealing temperature in the range of $700\text{--}850^\circ\text{C}$. These data suggest that YBCO in its crystalline structure is more stable than amorphous YBCO. With tunnel barriers that are at least as chemically stable as sapphire, processing temperatures of approximately 800°C might be usable to crystallize a high- T_c counterelectrode in a flash anneal without degrading the properties of the barrier or the superconductor. Further work in this area will be directed toward more stable insulators than Al_2O_3 .

4.4 EPITAXIAL FILMS AND LAYERED STRUCTURES

The best substrates for epitaxial growth of YBCO films have been found to be $\text{SrTiO}_3(100)$ or (110) , followed by $\text{MgO}(100)$ and yttria-stabilized zirconia (YSZ). Since all of these surfaces have a higher symmetry than YBCO, epitaxial YBCO films tend to grow in a mosaic pattern with approximately half of the grains rotated precisely 90° from the rest. X-ray diffraction and transmission electron microscopy have provided details of the mosaic structure of epitaxial YBCO films crystallized from the amorphous state on SrTiO_3 substrates. The most

important result of the TEM characterization - presented at the March APS Meeting - was the confirmation of the surface composition obtained from XPS and Auger spectroscopy, crystal orientation obtained by x-ray diffraction, and substrate/film interface composition obtained from Auger depth profiles.

The epitaxial growth of YBCO films crystallized from the vapor phase has been monitored as a function of film thickness by *in-situ* RHEED measurements. Evidence for crystallinity and epitaxy has been found for YBCO films 10 nm thick. This observation is consistent with Auger depth profile measurements of the thickness of the substrate/film reacted layer and is expected to correlate with low microwave losses from the substrate/film interface. Data from the ultrathin YBCO films will be published in the SPIE conference proceedings.

4.5 TUNNELING INTO HIGH T_c OXIDE FILMS

As was reported in the 1988 Annual Report, the initial attempts made to form S-I-S tunnel junctions by depositing Pb or Nb cross-strips on YBCO produced junctions which had very high resistance and ohmic I-V curves. This was attributed to the thick non-superconducting surface layer of the superconductor. As the result of modifications of the procedures used to prepare YBCO films (see Section 4.2) films are being grown in which the degraded surface layer has been greatly reduced. Using these films, tunnel junctions were prepared with MgO as the barrier layer and Nb as the counterelectrode. In order to protect the surface, a thin (3 nm) Au proximity layer was evaporated on the YBCO. The gold layer, the barrier, and the Nb counterelectrode were all deposited at room temperature. In some of these junctions, superconducting shorts were observed between the two electrodes. This was an indication that the barrier layer was "patchy". Although disappointing with respect to the primary goal of producing a working tunnel junction, this result was important in that it provided non-ambiguous evidence that the new growth procedures had indeed prepared films which were superconducting up to the surface. These preliminary tunneling experiments were described in a presentation at

the March APS Meeting. Other junctions were also prepared which did not contain shorts. However, in those cases, the junctions exhibited resistive behavior. It has thus far not been possible to prepare junctions which have barriers that are continuous and are of the proper thickness to permit tunneling to occur. All of the results of efforts to fabricate YBCO tunnel junctions will be reported at the upcoming Applied Superconductivity Conference.

4.6 NEW FABRICATION AND DEVICE CONCEPTS

One potential important use for the new high- T_c oxide superconductors such as YBCO is in microwave applications. The ideal substrate material for this application is sapphire because of its low dielectric losses. As has been discussed, sapphire, unfortunately, is not a good substrate for the growth of YBCO due to its relatively high reactivity with YBCO. On the other hand, the substrates that are currently being used for the epitaxial growth of YBCO would not be desirable. The dielectric losses are much too high in SrTiO_3 and only marginally acceptable in MgO and YSZ. A novel solution, for which a patent application is being filed, is to grow a thin epitaxial layer of MgO on sapphire which would then serve as the substrate for the epitaxial growth of YBCO. Three different orientations of MgO have been grown as single crystal films on different faces of sapphire. The epitaxial relationships were found to be the same as those reported for NbN grown on sapphire: $\text{MgO}(100) \parallel \alpha\text{-Al}_2\text{O}_3(1\bar{1}02)$, $(110) \parallel (10\bar{1}0)$, and $(111) \parallel (11\bar{2}0)$.

Measurements have begun on superconducting resonator cavities using epitaxial films grown on standard substrates. The first films grown on SrTiO_3 with an a-axis growth direction, had a surface resistance of 10^{-3} ohm/square at 2.8 GHz. On films using epitaxial MgO on sapphire as substrates, measurements will be made for both the free surface of the film and the surface adjacent to the substrate. The substrate/film interface is equally important as the free surface since total losses will likely be determined by both contributions. This work will be presented at the Applied Superconductivity Conference.

A new method for fabricating contacts between superconducting stripline interconnects and semiconductor devices has been conceived and a patent disclosure written and submitted.

5. PUBLICATIONS

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2. "Near-Surface Atomic Segregation in YBCO Thin Films," J. R. Gavaler, and A. I. Braginski, Physica C 153-155, 1435 (1988).
3. "Material Constraints on Electronic Applications of Oxide Superconductors," A. I. Braginski, Physica C 153-155, 1598 (1988).
4. "Crystal Lattice Measurements in Superconducting Materials," F. W. Lytle, E. C. Marques, R. B. Greegor, H. G. Ahlstrom, E. M. Larson, D. E. Peterson, and A. J. Panson, to be published in SPIE Proc. 879 (1988).
5. "Fabrication of High- T_c Superconducting $YBa_2Cu_3O_7$ Films," J. R. Gavaler, A. I. Braginski, J. Talvacchio, M. A. Janocko, M. G. Forrester, and J. Gregg, in MRS Vol. EA-14: High-Temperature Superconductors II, ed. by D. W. Capone II, W. H. Butler, B. Batlogg, and C. W. Chu (Mater. Res. Soc., Pittsburgh, 1988) pp. 193-196.
6. "In-Situ Fabrication, Processing, and Characterization of Superconducting Oxide Films," A. I. Braginski, J. Talvacchio, J. R. Gavaler, M. G. Forrester, and M. A. Janocko, in SPIE Proceedings Vol. 948, High- T_c Superconductivity: Thin Films and Devices, edited by R. B. van Dover and C. C. Chi (SPIE, Bellingham, Washington, 1988) - in press.
7. "Critical Currents of 85K Bi-Sr-Ca-Cu-O," G. R. Wagner, J. Talvacchio, and A. J. Panson, Mater. Lett. 6, 390 (1988).
8. "Prospects for Thin-Film Electronic Devices of High- T_c Superconductors," A. I. Braginski, M. G. Forrester, J. Talvacchio, and G. R. Wagner, Proc. 5th Intl. Workshop on Future Electron Devices (Miyagi-Zao, Japan 1988), pp. 171-179.
9. "Electrical Contact to Superconductors," J. Talvacchio, to be published in IEEE Trans. CHMT (1988).

*Submitted for publication in 1987.

10. "'MBE' of Superconducting Materials," A. I. Braginski and J. Talvacchio, to appear in Modern Superconducting Devices ed. by S. T. Ruggiero and D. A. Rudman (Academic Press, New York, 1989), in preparation.

6. PERSONNEL

A. I. Braginski	}	Principal Co-Investigators
J. R. Gavalier		
J. Talvacchio		

J. Gregg
M. A. Janocko
A. J. Panson
S. Sinharoy

7. COUPLING ACTIVITIES*

1. "Near-Surface Atomic Segregation in YBCO Thin Films," J. R. Gavaler, and A. I. Braginski, Contributed presentation at High- T_c Superconductors and Materials and Mechanisms of Superconductivity, Interlaken, February, 1988.
2. "Material Constraints on Electronic Applications of Oxide Superconductors," A. I. Braginski, Invited presentation at High- T_c Superconductors and Materials and Mechanisms of Superconductivity, Interlaken, February, 1988.
3. "In-Situ Fabrication, Processing, and Characterization of Superconducting Oxide Films," J. Talvacchio, A. I. Braginski, J. R. Gavaler, M. A. Janocko, and M. G. Forrester, Invited presentation to SPIE Meeting, Newport Beach, March, 1988.
4. "Near-Surface Composition and Properties of YBCO Films," J. R. Gavaler, M. A. Janocko, J. Talvacchio, M. G. Forrester, and A. I. Braginski, Contributed presentation at the March Mtg. of the American Physical Society, New Orleans, March, 1988.
5. "Microstructural Investigation of Epitaxial YBCO Films," J. Gregg, J. Talvacchio, J. R. Gavaler, and A. I. Braginski, Contributed presentation at the March Mtg. of the American Physical Society, New Orleans, March, 1988.
6. "SrTiO₃ Substrate Preparation and Nucleation of YBa₂Cu₃O₇ Thin Films," J. Talvacchio, Contributed presentation at the March Mtg. of the American Physical Society, New Orleans, March, 1988.
7. "Critical Currents of 85K Bi-Sr-Ca-Cu-O," A. J. Panson, G. R. Wagner, and J. Talvacchio, Contributed presentation at the March Mtg. of the American Physical Society, New Orleans, March, 1988.
8. "Fabrication of High- T_c Superconducting YBa₂Cu₃O₇ Films," J. R. Gavaler, A. I. Braginski, J. Talvacchio, M. A. Janocko, M. G. Forrester, and J. Gregg, Contributed presentation to the Materials Research Society Spring Meeting, Reno, April, 1988.

*Speaker's name is underlined.

9. "Epitaxial Growth and In-Situ Analysis of Thin Films for Superconductive Electronics," J. Talvacchio, Seminar at IBM T. J. Watson Research Center, Yorktown Heights, May, 1988.
10. "Prospects for Thin-Film Electronic Devices of High- T_c Superconductors," A. I. Braginski, M. G. Forrester, J. Talvacchio, and G. R. Wagner, Invited presentation to the 5th Intl. Workshop on Future Electron Devices, Miyagi-Zao, Japan, June 1988.
11. "Mechanisms of J_c Limitation in High- T_c Oxide Superconductors," A. I. Braginski, Invited panel presentation to the 5th Intl. Workshop on Future Electron Devices, Miyagi-Zao, Japan, June 1988.
12. "Progress Toward Electronic Applications of High- T_c Superconductors," A. I. Braginski, Invited presentation to the Materials Research Society International Meeting on Advanced Materials, Tokyo, June 1988.
13. "Electronic Applications of High- T_c Superconductors," A. I. Braginski, Seminars at Hitachi CRL, Sumitomo, Matsushita, Fujitsu-Atsugi, and NTT Ibaraki Laboratories, June, 1988.

*Speaker's name is underlined.

8. PATENTS

1. J. Talvacchio, "Epitaxial Buffer Layers for Growth of Oxide Superconductors on Sapphire," Disclosure No. RDS-88-046.
2. J. Talvacchio, "Method for Making High-Current, Ohmic Contact between Semiconductors and Oxide Superconductors," Disclosure No. RDS-88-057.